PB86-218120

Conceptual Design and Chemical Data Needs for Evaluation of Regional Acidic Deposition Models

(U.S.) Environmental Protection Agency
Research Triangle Park, NC

Jul 86

U.S. Department of Commerce
National Technical Information Service
NTIS

PB86-218120

EPA/600/D-86/138
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CONCEPTUAL DESIGN AND CHEMICAL DATA NEEDS FOR EVALUATION OF REGIONAL ACIDIC DEPOSITION MODELS

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TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
EPA/600/D-86/138		PB86 218120/AS

4. TITLE AND SUBTITLE	5. REPORT DATE
CONCEPTUAL DESIGN AND CHEMICAL DATA NEEDS FOR EVALUATION OF REGIONAL ACIDIC DEPOSITION MODELS	July 1986
	6. PERFORMING ORGANIZATION CODE
	-
7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
R.L. Dennis, M.C. Dodge and S.K. Seilkop	
9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
(Same as Block 12)	CCVNIA/05-5104 (FY-86)
	11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Atmospheric Sciences Research Laboratory--RTP, NC Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, North Carolina 27711	14. SPONSORING AGENCY CODE EPA/600/9

15. SUPPLEMENTARY NOTES

16. ABSTRACT

The system behavior of regional models can not be evaluated with existing data bases, such as SURE, OSCAR, or the NTN/NADP, in part because they were not designed with model evaluation in mind. Simultaneous measurements of air quality, precipitation chemistry, and dry deposition, which are necessary for a fully rigorous integrated evaluation, are lacking in present data bases. New, more comprehensive data sets with coincident measurement of the major species are needed. A discussion of data needs for the collection of chemical data as part of the design of a new field study to collect such data is presented. Since a surface network will be the backbone of the data base, the chemical data needs of the surface network receives the most attention. First the species which need to be measured are discussed, and then the spatial and temporal data needs are discussed.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
RELEASE TO THE PUBLIC	UNCLASSIFIED	
	20. SECURITY CLASS (This page) UNCLASSIFIED	22. PRICE

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE

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INTRODUCTION

A major application of regional acidic deposition models is to estimate changes in patterns of deposition due to changes in emissions. To estimate these patterns and changes in them the models must be capable of estimating deposition loadings with the proper source attribution while accounting for chemical nonlinearities. The changes are not necessarily expected to be obvious due to anticipated nonlinearities resulting from competition among chemistry cycles. Because the physical-chemical processes are complex, models are important as a tool for investigating acid deposition and formulating control policy. For policy use model predictions need to be credible.

For a model to be credible several conditions must be met: (1) the most important processes must be included in the modeling framework, (2) incorporation of these processes into the model must be adequate, and (3) the model system as a whole must perform adequately. An assessment of (3) and aspects of (2) can be achieved by comparing the predictions of a model and its modules (components) against real-world observations in a rigorous model evaluation. Such an assessment allows one to develop confidence that the model is producing correct answers for the right reason. Confidence in the models is important because they are required to make predictions for conditions significantly different from today's conditions. A model evaluation aimed at assessing the description of individual processes or modules of a model can be termed a mechanistic evaluation. One aimed at assessing the system performance with respect to predicting deposition at the surface can be termed an integrated evaluation. Both need to include diagnostic elements to develop confidence and credibility. In this paper we address the data needs for an integrated evaluation only. Also, for sake of brevity, we treat in detail only those data needs related to the collection of chemical data for an integrated evaluation.

The system behavior of regional models can not be adequately evaluated with existing data bases, such as SURE, OSCAR, or the NTN/NADP, in part because they were not designed with model evaluation in mind. Simultaneous measurements of air quality, precipitation chemistry, and dry deposition, which are necessary for a fully rigorous integrated evaluation, are lacking in present data bases. New, more comprehensive data sets with coincident measurements of the major species are needed. In this paper we present a discussion of data needs for the collection of chemical data as part of the design of a new field study to collect such data.

DATA BASE CHARACTERISTICS FOR AN OPERATIONAL EVALUATION

New data bases for model evaluation should address the comprehensive Eulerian regional models presently being developed. They should include measurements which allow the definition of the patterns of output variables and which allow diagnosis of those patterns. Considering characteristics of acid deposition and of model computations, requisit features of an evaluation program are:

- the temporal scale of diagnostic importance is event-related.
- the temporal scale of user interest is seasonally-related.
- chemical detail is vital.
- Simultaneous measurements of air and aqueous chemical quality is crucial.
- 3-D features of concentration fields is of diagnostic importance.

In addition, subgrid-scale processes are important. The grid resolution of the Eulerian models is coarse relative to the spatial scales that govern processes such as chemical mixing and scavaging in cumulus clouds. The effect of averaging over 80120 km grid cells must be understood. Also, it appears that individual monitoring sites are not regionally representative with respect to deposition fields, especially for wet deposition. Characterizing the uncertainty in the observed regional deposition fields, requiring subgrid-scale information, is important.

From the above discussion four components of a single data base which supports an integrated evaluation are implied: (a) a surface monitoring network to collect a full suite of chemical data related to wet and dry processess and to establish regional patterns of deposition, (b) measurements in the volume above the network, (c) measurements of transport and (d) subgrid scale measurements. The measurements will need to spatially and temporally resolve deposition events. For the large regional scales being addressed by the models, the surface network will be the backbone of the data base. Aircraft availability is limited and spirals and transects are expensive as are dense surface networks. Thus we concentrate on the chemical data needs of the suface network. We will first discuss which species need to be measured. Then we will discuss the spatial and temporal data needs.

CHEMICAL DATA NEEDS

The regional models under development contain a reasonably detailed description of the gas- and aqueous-phase chemical transformations that occur in the atmosphere. The gas-phase mechanisms in these models describe how free radicals and other oxidizing species that are formed in the photochemical process convert SO2 to sulfate and NOx to nitric acid. These acids can be neutralized, dry deposited or scavenged by cloudwater droplets. In addition to the acids, other chemical species in the gas-phase mechanism, such as SO2, HOOH, and O3, can also be absorbed into cloud droplets and undergo chemical conversions there that lead to acid formation. The clouds can either precipitate, resulting in the wet deposition of acidic material, or they can evaporate, leaving behind sulfate and nitrate aerosols that eventually may be dry deposited or reincorporated into clouds.

The formation of acids in the environment depends on many factors including (1) the amount of acid precursors emitted into the atmosphere, (2) the chemical reactions that convert the precursors into acids, (3) solar radiation and atmospheric variables, which influence the rate of

conversion of SO2 and NOx to sulfate and nitrate, and (4) dispersion, which determines where and in what amounts the acids will be deposited. In order to determine if all these complex and interrelated processes are accurately represented in the models, an array of different chemical species should be measured during the field program. The required gas- and particulate-phase species are given in Table I; the aqueous-phase species that should be measured are given in Table II. Measurements of this array of species is designed to unravel the source of any discrepancies between predictions and observations.

Each species listed in Tables I and II can be characterized as being of one or more of the following types:

Input Variables:	Species that are emitted into the atmosphere
Output Variables:	Species that result in acidification of the environment when dry or wet deposited
Diagnostic Variables:	Species that serve as a check on model performance and can be used to trace cause-effect linkages

Some of the species involved in the acidification process, such as SO2, serve dual roles. SO2 is both an input variable and also an output variable since any SO2 that is dry deposited is ultimately converted to acid. The output variables that contribute directly to acidity in the environment and, therefore, must be measured to evaluate the overall performance of the models are HNO3 and particulate sulfate and nitrate in the ambient air and sulfate, nitrate, and pH (H+) in rainwater. These can be considered to be the major species of concern. The rationale for measuring the other species listed in the tables is as follows:

NO, HC, and CO. All three species are input variables. Measurements of NO and HC in source areas can serve as a check on the accuracy of the emission inventory. Measurements of HC aloft are especially important for establishing initial and boundary conditions for the models. Carbon monoxide, which is a conservative species, is a tracer for air masses and can serve as a partial check on the treatment of transport in the models.

NO2 and O3. Both species can be defined as diagnostic and output variables. They are both important products in the photochemical cycle and, as such, they can serve as diagnostic checks on how well chemical transformations are being represented in the models. Ozone can be absorbed by cloud droplets and react there with dissolved SO2 to form sulfate. NO2 in the gas phase reacts with OH radicals to generate HNO3. The dry deposition of O3, which is a powerful oxidant, can lead to further acidification of the environment. The dry deposition of NO2 can also have an adverse effect on the terrestrial ecosystem by making it more susceptible to damage by acids.

PAN. Peroxyacetyl nitrate can be defined as both a diagnostic and an output variable. PAN can thermally dissociate to NO2 and thereby provide an additional route to HNO3 formation in the atmosphere. The radicals that are produced along with NO2 in this thermal dissociation process can also

accelerate the formation of O3 in the atmosphere and lead to an increase in the rate of conversion of SO2 to SO4= and NO to NO3-. It is also expected that PAN, once it is dry deposited, will readily be converted to nitrate and thereby increase acidity in the terrestrial ecosystem.

HCHO. Formaldehyde can be considered to be a diagnostic and an output variable. This extremely reactive species, which is formed during the oxidation of hydrocarbons, is a major source of free radicals in the atmosphere. The free radicals generated from HCHO can lead to the gas-phase oxidation of SO2 and NOx. Formaldehyde can also be scavenged by cloudwater droplets. Once in the aqueous-phase, HCHO can react with dissolved SO2 to form a complex that is resistant to oxidation. Formaldehyde also can be deposited in aquatic ecosystems where it can oxidize the natural sulfur that is found in surface waters.

HOOH. Hydrogen peroxide is both a diagnostic and an output variable. This species is potentially the major oxidant for the conversion of dissolved SO2 to sulfate in cloudwater droplets. The wet and dry deposition of this powerful oxidant can also lead to increased acidification of aquatic systems since it will readily convert SO2 to sulfate in surface waters.

NH3 and NH4+. Ammonia, which is an input variable, is readily converted to ammonium ion in cloud and rainwater. NH4+ is an important output variable because it is the major neutralizer of acids in cloud and rainwater.

Fe+3, Mn+2, Ca+2, Mg+2, Cl-. All five of these ions can be considered to be both input and output variables. Iron and manganese, which are emitted from industrial areas, catalyze the air oxidation of SO2 to sulfate in cloudwater. Calcium and magnesium, which are found in soil dust and Cl-, which is a component of sea salt, can influence the acidity of rainwater. Measurements of these five species are needed to establish ion balances and serve as a check on the pH measurements.

The species were selected in an attempt to weigh coverage versus detail. A regional network cannot be run as a research-grade set of sites; it must be operated as a routine network. This consideration sets limits on the number of species that can be routinely monitored; the sub-set shown in the tables for a routine network includes only those species that are considered to be essential for an overall evaluation of the models. It is recommended that coincidental measurements for the full array of species be carried out at only five to ten special sites.

Aircraft measurements should be conducted on a periodic basis to determine the vertical and horizontal distribution of pollutants. Vertical profiles are especially necessary to provide initial and boundary conditions for comparison with the model predictions. Vertical resolution above the ground-based network is also needed to determine the extent to which vertical fluctuations in ambient concentrations affect predictions of wet and dry deposition at the surface. The chemical species that should be measured during these periodic aircraft flights are listed in Table 1. To the extent possible, the aerial sampling program should include measurements of the same species that will be measured at the surface network sites. Because of the importance of establishing initial and

boundary conditions, measurements of reactive organics aloft also should be made. Grab samples should be collected for subsequent speciated analyses of the hydrocarbons. In addition, preliminary sensitivity studies conducted with one of the regional models under development indicate that predictions of wet deposition are strongly dependent on the assumed concentration of formaldehyde aloft. Consequently, aerial measurements of this important free-radical initiator should also be made.

In addition to obtaining vertical and horizontal pollutant profiles, a concerted effort should be undertaken to provide the data that are needed to evaluate the treatment of cloud chemistry in the regional models. Toward this end, measurements of the key species involved in aqueous-phase chemical transformation processes should be measured above, below and within clouds. The required ambient measurements at cloud base and cloud top are listed in Table I. The needed cloudwater measurements are given in Table II. Those species that lead to the oxidation of SO2 within clouds, namely HOOH and O3 are especially important to measure. In addition, ambient measurements of HCHO are desirable since in cloudwater this species rapidly forms a complex with SO2 that is highly resistant to oxidation. Accurate gas-phase and cloudwater aerial measurements of HOOH are especially needed to provide the data necessary to evaluate aqueous-phase chemistry in the regional models.

In addition to listing what species need to be measured and where they should be measured, Tables I and II also list the individual components of the models whose performance can be evaluated if this data base is obtained. The individual model components include emissions, transport, gas- and aqueous-phase chemistry, wet deposition and dry deposition. Acquiring data for many of the species will enable one to assess the reliability of a number of different model components. For example, since HOOH is a product of photochemical smog, measurements of this species will provide an indication of how well chemical processes are treated in the models. Since HOOH is the major oxidizer of SO2 in cloudwater, aerial measurements of this species above, below and within clouds will enable one to assess how well wet deposition is handled in the models. Measurements of HOOH at ground level will permit an assessment of the models' ability to accurately treat the role of this powerful oxidant in dry deposition.

SPATIAL DATA NEEDS FOR SURFACE NETWORK

This section addresses the spatial requirements of the surface network as they relate to comparisons of model predictions and observations. Criteria for network adequacy are presented based on uncertainty limits about identified patterns of regional deposition. Using currently available data, we examine a proposed network's ability to satisfy these criteria, and explore its ability to discriminate and define regional patterns.

In performing an integrated evaluation, the output fields of the model will be compared with observations. Output fields of major interest are regional patterns of deposition of sulfate, nitrate and hydrogen ion. For dry deposition the comparison will need to be indirect, since dry deposition is generally inferred from surface ambient air measurements using appropriate deposition velocities to obtain dry deposition fluxes.

Thus the evaluation of dry deposition will stress the comparison of predicted ambient concentration fields against observations.

NADP wet deposition data and SURE air concentration data in the northeast show evidence of random variations superimposed on the large-scale deposition and air concentration patterns. The implication is that monitoring sites are probably not regionally representative, i.e., not always equal to deposition or concentrations averaged over 100 x 100 kms. Model predictions, on the other hand, are regionally representative because inputs and processes are smoothed by the use of large grids cells (80 km or more on a side). Thus, a model evaluation should use spatial patterns estimated from observational data as a basis of comparison rather than individual site values.

For the comparison to be interpretable, the patterns should have commmensurate spatial scales of averaging. This necessitates smoothing or filtering the observational data to a degree consistent with the model predictions. Because of the random variation in observational data, a smoothed spatial pattern estimated from them has a component of uncertainty associated with it. Just as a sample mean imperfectly estimates the population mean, the estimated pattern imperfectly represents the true pattern. In performing a model evaluation, it is important to account for the error in the estimated pattern.

A spatial pattern representing model output can not be expected to conform precisely to that inferred from data. It should, however, lie within the uncertainty in the data's spatial pattern. For the comparison between model and data patterns to be rigorous, the uncertainty in the spatial data pattern must be within acceptable limits. If the uncertainty is too great, the evaluation will not be sensitive to a model's departure from reality. In designing the surface network, it is therefore essential to define criteria for acceptable limits of uncertainty in spatial patterns which can be estimated from the network. The following are operational criteria which provide a reasonable degree of sensitivity for model evaluation:

> The relative uncertainty in seasonal estimates from the network should not exceed +/-60%.
>
> In the regions of highest deposition or concentration, relative uncertainty in estimates should not exceed +/-30%.

An interpolation method known as kriging is used to evaluate a proposed surface network against these criteria. Kriging produces a "regression-like" smoothed estimate of the surface passing through spatial or geographic variables. It also produces an estimate of the uncertainty (expected squared error) of the interpolated points. Thus it can be used to place confidence bands about the estimated spatial patterns. This later feature makes kriging unique among interpolation schemes. The kriging estimator is a best linear unbiased estimator, that is, it gives no systematic error (bias) and has the smallest estimation variance among unbiased estimators that are linear functions of the data. Kriging is based on Matheron's theory of regionalized variables (Matheron, 1965, 1973), which was designed to mathematically describe phenomena distributed in space

and/or time and provide an appropriate means for solving spatial estimation problems. For a general discussion of kriging, see Delhomme (1978).

The proposed surface network is composed of the first 35 NTN dry deposition sites to be placed in the northeast, the present UAPS network, 5 stations from a new New York network and 11 stations from Canada, for a total of 70 stations. The network design, which incorporates reasonably uniform spacing, is shown in Figure 1. For our spatial design analysis, currently available data are used to define a prototypal spatial covariance structure for the variable of interest. This covariance structure is used in combination with the relative geometry of the proposed network sites to estimate the expected size of the variance about a spatial pattern which would be derived from the proposed network. The same random variability about the spatial pattern as observed in current data is assumed. For a given covariance structure the spacing of sites is an important determinant of the size of the uncertainty bands.

Using seasonal and annual sulfate wet chemistry data for 1981 and 82 from the NADP network in the northeast the variability, as a function of location, was calculated for the proposed network. The variability was divided by the observed 1981 patterns and multiplied by two to produce an approximate relative 95% confidence interval, that is, a 95% uncertainty band in terms of percent. The results for annual and summer variances for sulfate wet chemistry are shown in Figures 2 and 3, respectively.

These uncertainty estimates are based on individual point variances. As such, they are overestimates of the uncertainty about the surface estimate. The estimated surface is composed of individual points with individual variances just as a sample mean is a function of individual values and variances. Like the variance of a sample mean, the variance of the estimate surface is not accurately represented by variances associated with its individual values. The individual variances are larger than the variance of the sample mean. Kriging uncertainty is the uncertainty associated with each of the point estimates or individual variances. Thus it provides an upper bound for the uncertainty in the mean of the surface estimate. We must be aware of this fact in making our assessments.

For annual sulfate wet deposition the two standard deviation relative uncertainty interval is +/-40% over the major region of deposition. It is smaller for rainwater concentration, being +/-30%. For seasonal wet deposition the relative uncertainty interval is the same across the seasons, being 50-60%. However, the rainwater concentration uncertainty ranges from 30% for summer to 60% for winter. (The variances are similar, but the mean concentration is higher in summer.) Hence, the 70 station network will meet our more stringent criterion of 30% for annual wet chemistry patterns and may meet it or come close for seasonal. We can't be sure about the seasonal until the error about the surface estimate is directly estimated.

Similarly, the SURE data set was used to develop the variance estimates for particulate sulfate patterns. Figure 4 shows the uncertainty estimate for July 1978. The relative uncertainty is +/-50% over the area of major SO4 concentration. Again, this is an overestimate because it is based on individual point variances. Combining four months, one in each season produced confidence bands of +/-40% over the region and +/-30% over

the central area of highest concentration. Thus we expect the network will meet our more stringent criterion for annual but may not for seasonal particulate sulfate patterns. However, the network may not be dense enough to meet our criterion for monthly averages of particulate sulfate.

Individual episodes will also need to be examined. Clearly they will show much larger error bands than the seasonal results presented. We will also examine the sensitivity of the error variance to station density. After sulfate we will examine the determination of spatial patterns of SO2, but we suspect the network will not be capable of reliabley defining its regional pattern due to subnetwork spatial structure.

Similar design analysis using present data bases, such as the OSCAR high density network, needs to be performed for the planning of subgrid scale monitoring. The conceptual design process can use kriging as a technique. Work is underway to this effect.

Placement of the special sites among the planned network will have to be assessed through model sensitity studies. The model can be used as a laboratory to test chemical non-linearities that should influence surface measurements. The model runs would also investigate the need for aircraft flights in conjunction with the special surface sites.

TEMPORAL DATA NEEDS FOR SURFACE NETWORK

An important criterion for purposes of model evaluation is the ability to temporally resolve events. The model evaluation will depend on comparison of individual cases and the ability to define the events making up the seasonal averages. Three techniques were used to develop preliminary recommendations for temporal data needs for ambient concentration measurements. They were graphical analysis, autocorrelation analysis and cross-correlation analysis. A fourth technique, fourier analysis, was investigated, but its results were not felt to be sufficiently reliable because patterns of concentrations are not that periodic, although conclusions from it were consistent with the other three techniques.

Data from several sites were used in the analyses: a research site at Argonne, IL (SO2), and the SURE Class 1 sites of Montague, MA, Duncan Fall, OH and Ft. Wayne, IN (SO2, NOx and sulfate). Temporal plots showed that broad-scale peaks in concentrations had time scales on the order of a day or more, but were aperiodic in their occurance. Hourly values for SO2 and NOx and 3-hourly values for sulfate were averaged to produce time series of 3-, 6-, 12-, and 24-hour averages.

The analyses indicated that for averaging periods up to six hours the temporal events continued to be reasonably well represented, although there is some loss in precision in resolving events in going from 3-hour to 6-hour averages. Twelve-hour averages are just barely adequate to represent the temporal events and 24-hour averages lose much of the information about the events. For example, the data are strongly autocorrelated. The degree of autocorrelation remained strong for 6-hour averages, was weak for 12-hour averages and was no longer statistically significant for 24-hour averages.

There are also temporal needs from the point of view of chemistry. Because chemistry is influenced by the diurnal cycle of sunlight, the chemical data bases collected must be able to resolve temporally daily episodes. Many species slated for measurement are expected to show significant diurnal variation. From that standpoint it is recommended that the temporal resolution of the measurements for all ambient air species with the exception of ozone be 3-6 hours in order to capture the essence of the diurnal cycle. Because of the significant diurnal variation of ozone, this species should be monitored on an hourly basis.

There are competing considerations for wet chemistry. Problems with data and the definition of an event lead to a recommendation for straight 24 hour sampling (Jake Hales, private communication). Trajectory analysis performed with Whiteface event data indicate that the time average of the precipitation event should be no more than 6 hours. Otherwise information as to the origin of the air mass which is being scavaged by the precipitation is lost (Billman Stunder, Heffter and Dayan, in press). We recommend, therefore, that there be special sites with 3-6 hour temporal resolution of the precipitation chemistry and that all or most of the sites be equiped with recording rain gauges to get temporal resolution of the precipitation.

SUMMARY

Chemical data needs to support an integrated model evaluation, combining operational and diagnostic needs, were examined. A suite of chemical species was identified which should concurrently be measured to support an integrated model evaluation. Not all species can currently be routinely monitored. Hence a surface network for those which can be and special sites for the entire suite is the design concept. For the routine surface sites on the order of 70 stations across the Northeastern U.S. and Canada are needed to identify regional patterns of deposition and concentration precisely enough for an integrated evaluation. To adequately identify event features the time resolution of the surface network needs to be no more than six hours.

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TABLE I: AEROMETRIC MEASUREMENT NEEDS

	Surface		Aloft		
Species	Special Sites	Regional Network	Vert/Horiz Profiles	Cloud Base/Top	Modules*
SO2	x	x	x	x	1,5
NO	x				1
NO2	x	x	x		3,5
PAN	x	x	x		3,5
HNO3	x	x	x	x	3,4,5
NH3	x	x	x	x	1
O3	x	x	x	x	3,4,5
HOOH	x			x	3,4,5
CO	x				1,2
HC	x		x		1,3
HCHO	x		x	x	3,4,5
SO4= (part.)	x	x	x	x	3,5
NO3- (part.)	x	x	x	x	3,5
NH4+ (part.)	x	x	x	x	5

*
Numbers refer to the modules whose performance can be evaluated if measurements for the corresponding species are obtained:

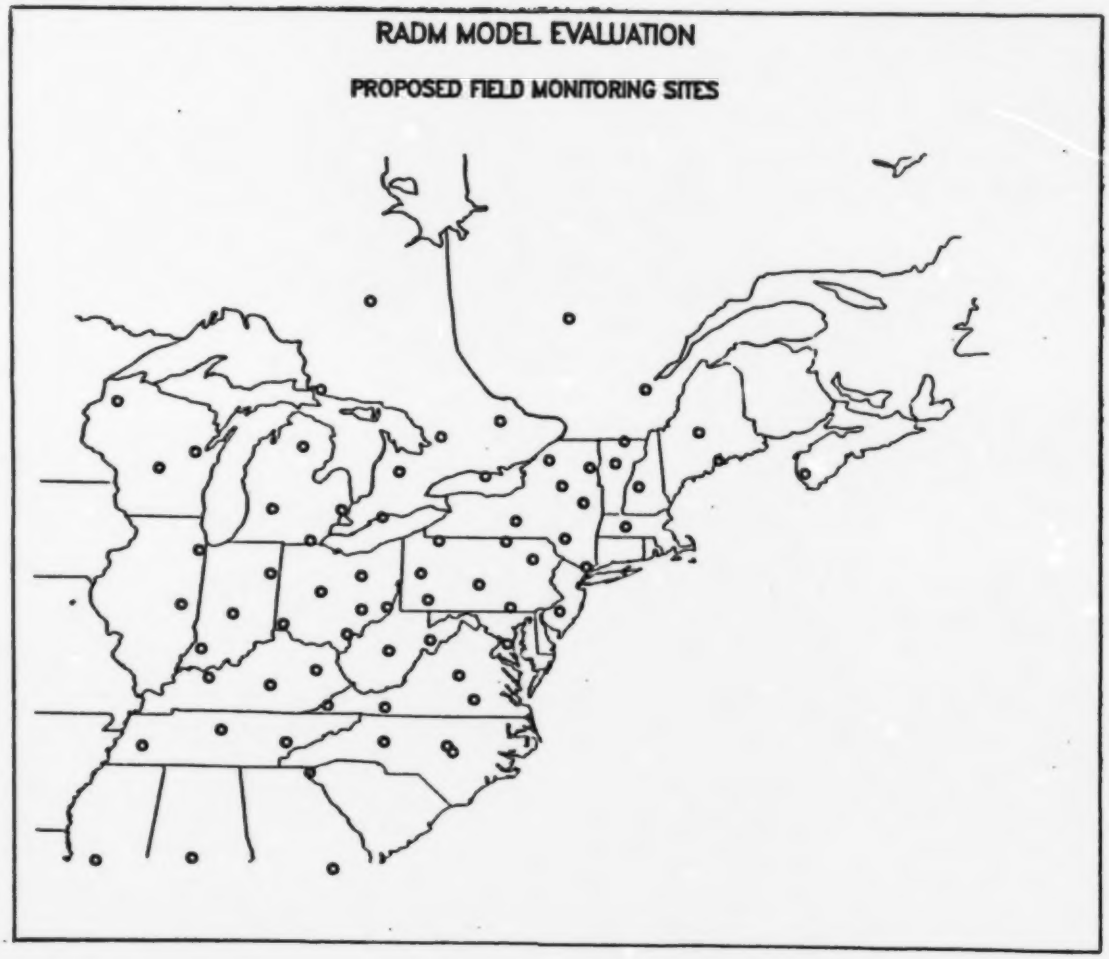
1. Emissions
2. Transport
3. Chemistry
4. Wet Deposition
5. Dry Deposition

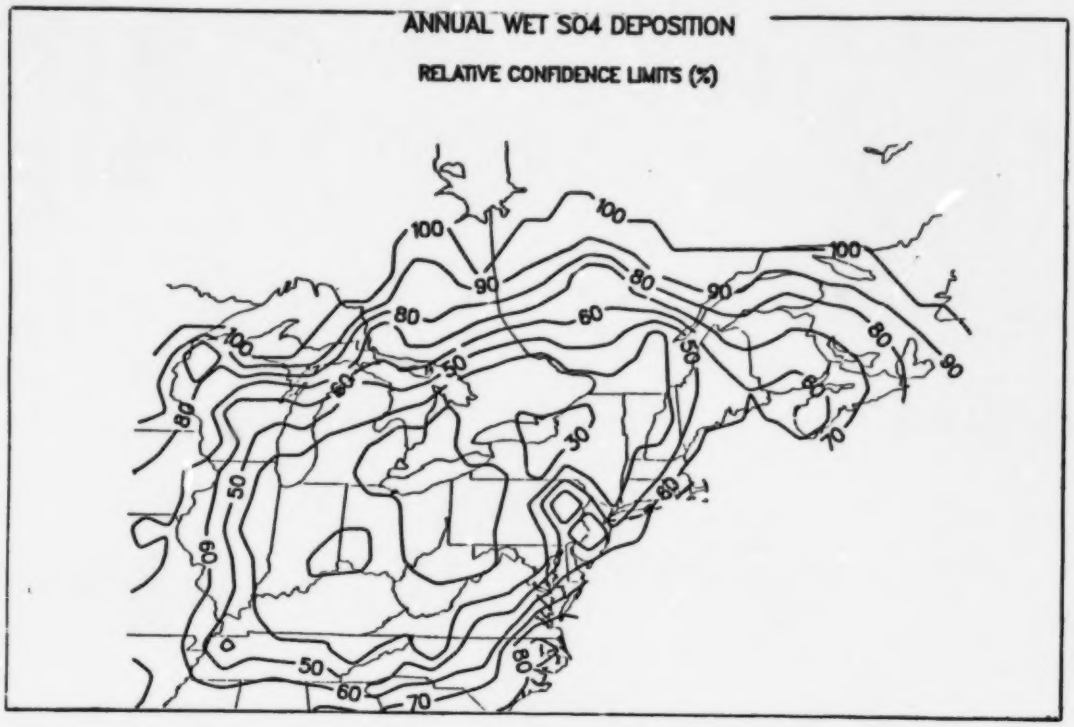
TABLE II: AQUEOUS MEASUREMENT NEEDS

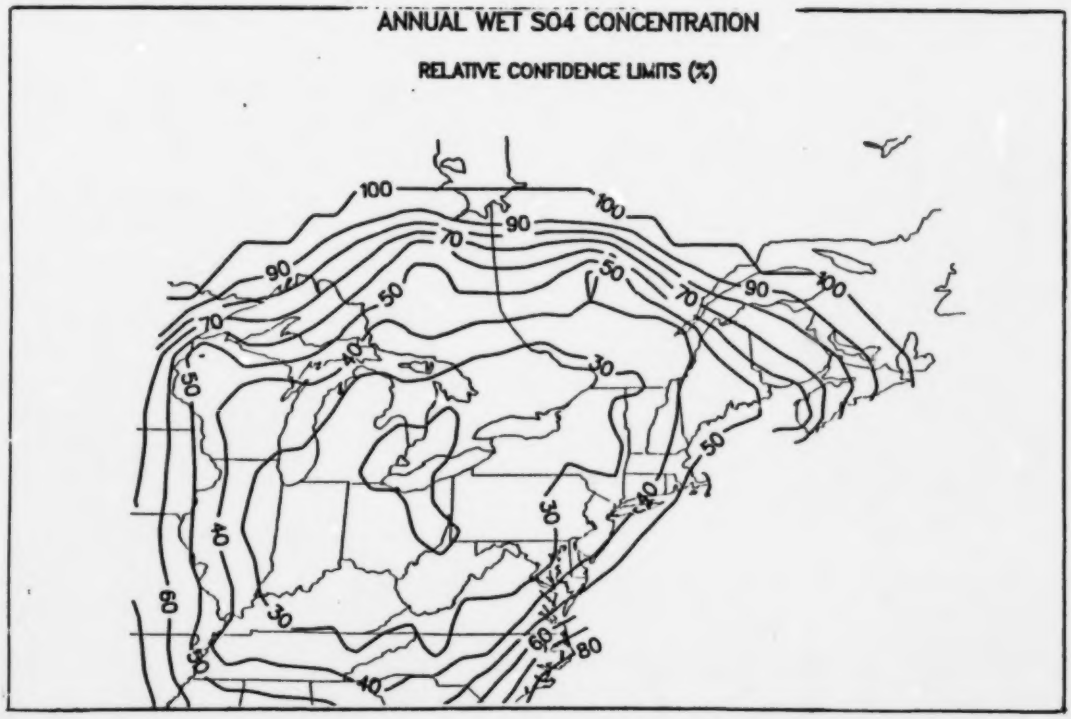
Species	Surface Rainwater: Special Sites	Surface Rainwater: Regional Network	Cloudwater	Modules*
SO4=	x	x	x	3,4
NO3-	x	x	x	3,4
NH4+	x	x	x	4
H+	x	x	x	4
Fe+3	x	x		1,4
Mn+2	x	x		1,4
Ca+2	x	x		4
Mg+2	x	x		4
Cl-	x	x		4
HOOH	x		x	3,4
HCHO	x		x	3,4

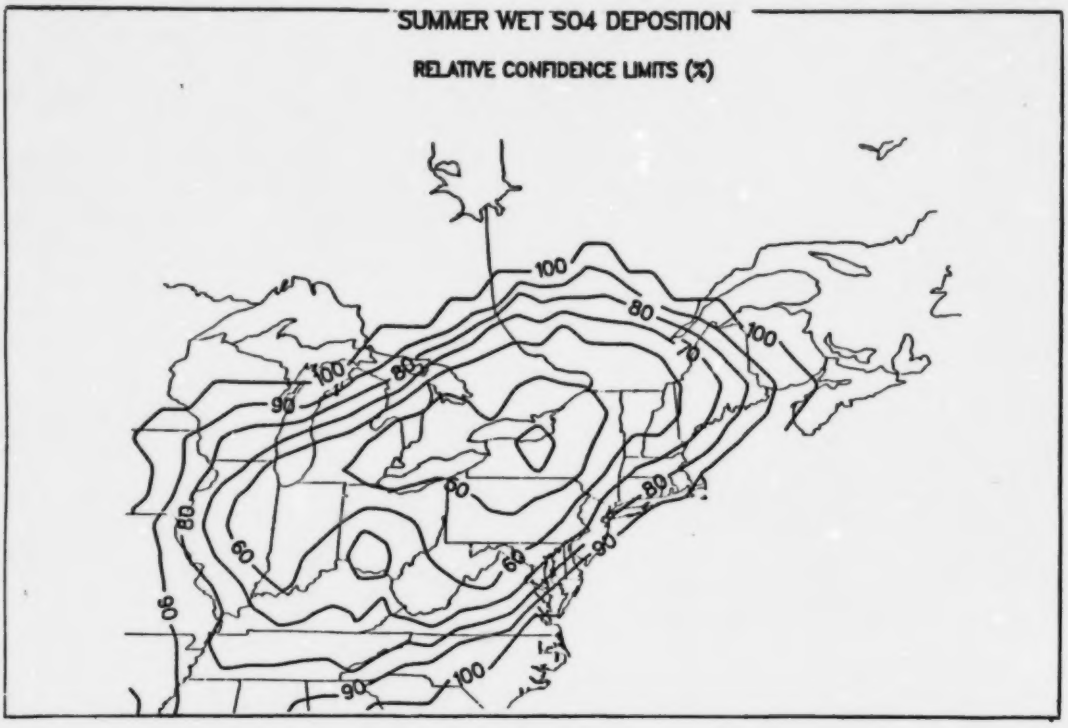
*
Numbers refer to the modules whose performance can be evaluated (see TABLE I):

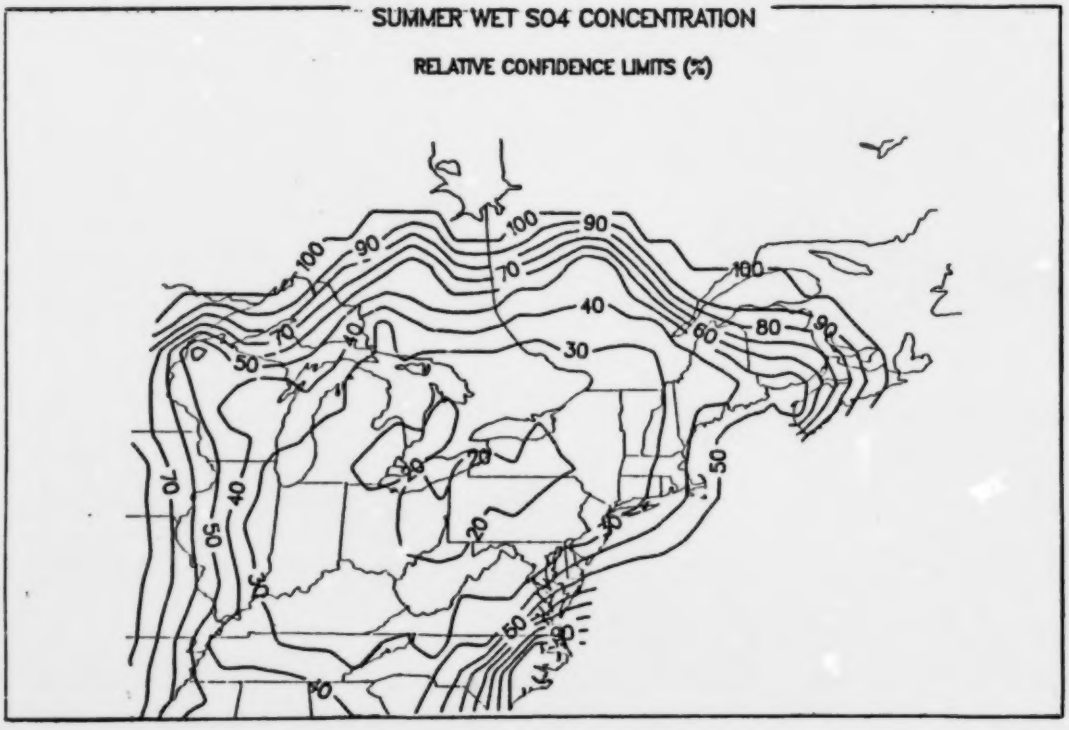
1. Emissions
3. Chemistry
4. Wet Deposition

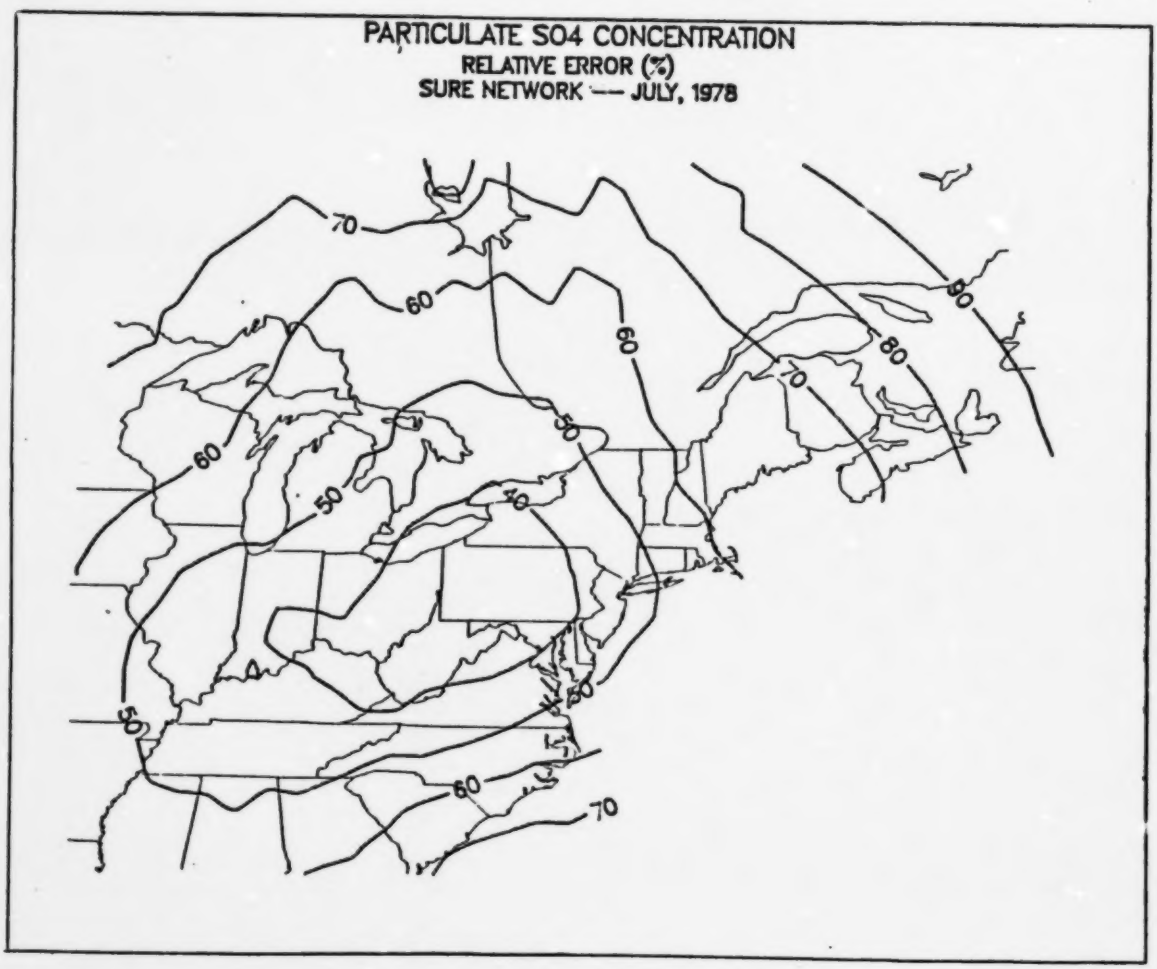

RADM MODEL EVALUATION
PROPOSED FIELD MONITORING SITES


ANNUAL WET SO4 DEPOSITION
RELATIVE CONFIDENCE LIMITS (%)


ANNUAL WET SO4 CONCENTRATION
RELATIVE CONFIDENCE LIMITS (%)


SUMMER WET SO4 DEPOSITION
RELATIVE CONFIDENCE LIMITS (%)
100
80
90
70
60


SUMMER WET SO4 CONCENTRATION
RELATIVE CONFIDENCE LIMITS (%)
100
90
70
40
30
80
60
50
20


PARTICULATE SO4 CONCENTRATION
RELATIVE ERROR (%)
SURE NETWORK — JULY, 1978
70
60
60
50
60
50
40
80
80
70
50
60
60
70